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High-Temperature Ceramic-Carbonate Dual-Phase Membrane Reactor for Pre-combustion Carbon Dioxide Capture – Process Design and Techno-Economic Analysis

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Abstract

Fossil fuels can be effectively converted to electricity by integrated gasification combined cycle (IGCC) technology, especially when carbon capture is required [1]. The process uses a high-pressure gasifier to convert coal into pressurized synthesis gas (syngas). The overall reaction/separation process for the IGCC power plant includes gasification, fly ash removal, 2 or 3 fixed-bed reactors for water-gas-shift (WGS) reaction (with at least two different catalysts), and H_2S and CO_2 removal by the low-temperature process such as methanol washing. WGS, an essential step in the IGCC process, is an exothermic reaction. Therefore, thermodynamically low reaction temperatures are desired. However, high temperatures are favorable from a kinetic viewpoint. Commercially, WGS is conducted in two or more reactor stages operated at different temperatures, with inter-cooling to maximize conversion for a given catalyst volume. The CO_2/H_2 stream from the reactors is subsequently separated by an amine absorption process. As shown in Figure 1(A), these reactions and separation processes are costly and energy-intensive [1].

Membrane reactors removing one of the products (i.e., H_2 or CO_2) from the WGS reaction zone can significantly enhance the CO conversion with simultaneous product separation. Our laboratory has recently significantly advanced in developing a high-temperature CO_2 perm-selective membrane reactor for the WGS reaction with CO_2 capture [1]. The membrane reactor employs a ceramic-carbonate dual-phase (CCDP) membrane perm-selective for CO_2 separation at high temperatures (600-900 °C). Combined with a high-temperature sulfur-resistant catalyst for WGS, the membrane reactor can achieve a >95% single-pass CO conversion with CO2 capture without requiring an additional downstream separation process, Figure 1(B) [3].

This paper presents the process design and techno-economic analysis (TEA) for a membrane reactor process with a CCDP membrane for the WGS reaction with CO_2 capture for a 550 MW coal-fired IGCC power plant and its comparison with the conventional fixed-bed reactor system with follow-up CO2 capture by the MEA process, as shown in Figure 1. Syngas from a GE Power (GEP) gasifier with Illinois 6# coal after purification is used as the feed to the membrane or conventional reactor. The target performance for the reactor for WGS with CO_2 capture includes CO conversion >95%, hydrogen stream purity >90%, CO_2 stream purity >95%, and total carbon capture >90% [1]. Using a high-temperature, sulfur-resistant commercial catalyst and a CCDP membrane developed in this project, the membrane reactor can achieve the performance target without a subsequent CO_2 capture process at 750 °C and space velocity of 250 h^{-1}, slightly larger than that for the conventional fixed-bed reactor on the same catalyst.

The membrane reactor in a structure resembling a shell-tube exchange consists of multiple cylindrical vessels, each containing many CCDP membrane tubes with WGS catalyst packed on the shell side. A mathematical model, verified with experimental data, is used to design the membrane reactor and its process with a given WGS catalyst and CCDP membrane.

The outcome of the process design and TEA analysis shows that the CCDP membrane reactor for WGS with in-situ CO_2 capture has an operating cost of $24M/year, significantly lower than that for the conventional fixed-bed reactor with a separate amine absorption process for CO_2 capture ($40M/year). However, the membrane reactor process has a much higher capital cost ($1,007M) than the conventional process ($527M) because of the higher cost of the CCDP membrane reactors ($951M) than that of the traditional reactors ($240M). Modeling analysis shows a membrane reactor using a CCDP membrane with higher CO2 permeance can deliver the targeted performance for WGS reaction with CO2 capture at a much higher space velocity and lower membrane surface area to catalyst volume ratio, leading to a smaller catalyst amount and/or membrane area and hence significantly reduced membrane reactor capital costs. TEA analysis of a membrane reactor using a CCDP membrane with a 2.4-fold increase in CO_2 permeance shows a reduction in the capital cost by 50%. The preliminary TEA shows the promise of the membrane reactor for WGS with in-situ CO_2 capture and suggests directions for further improvements to the membrane reactor process.

Figure and Tables

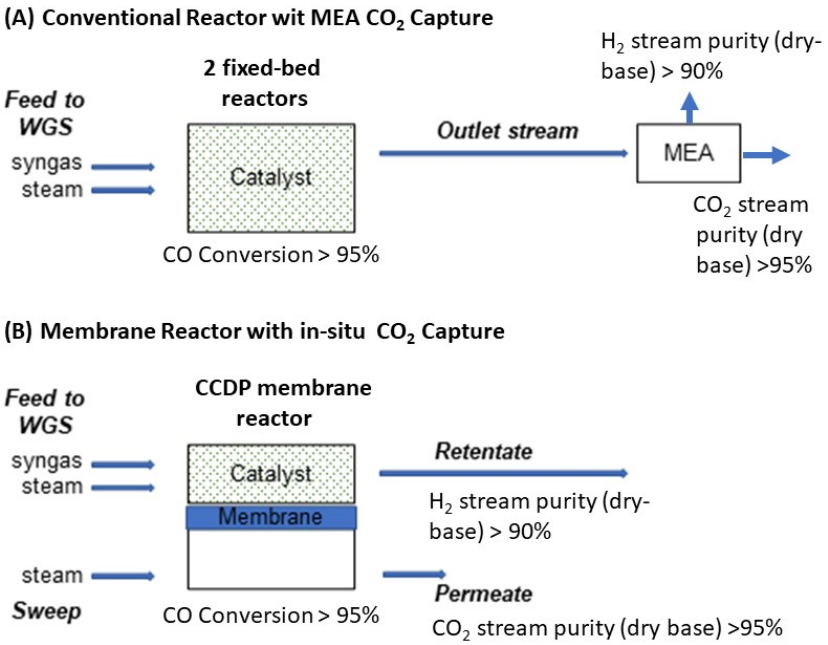

Figure 1. Schematic representation of conventional and membrane reactors for WGS reaction with CO2 capture.

References

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